

***RADIONUCLIDE MIGRATION IN PLUTONIC ROCKS:
IMPLICATIONS FOR HIGH-LEVEL NUCLEAR WASTE DISPOSAL.***

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This thesis contains no material which has been accepted for the award of any other higher degree or graduate diploma in any tertiary institution, and to the best of my knowledge and belief, no copy or paraphrase of material previously written or published by another person, except where due reference is provided.

A handwritten signature in black ink, reading "Vanessa Ann Guthrie". The signature is fluid and cursive, with a large, stylized 'V' and 'G'.

Vanessa Ann Guthrie.

February 1991.

*It is our responsibility to ensure that we leave for our children
the environment which we hold in trust not as a legacy
but as a protected and well managed resource.
Let us aim to meet the challenge.*

Abstract

Geochemical studies of deeply buried intrusive rocks provide the foundation for evaluating the suitability of crystalline rocks as repositories for solidified High-Level Nuclear Waste (HLW). This geochemical study examines the migration of natural and introduced radionuclides during interaction between groundwater and plutonic rock, to provide an understanding of the processes which may operate in the potential repository environment.

Rock cores from the Coles Bay Granite (Tasmania), Kambalda Granodiorite (Western Australia) and the Roxby Downs Granite (South Australia) were selected for this study on the basis of the variation in mineralogy, fracture density and degree of alteration. Firstly, the behaviour of U- and Th-decay series radionuclides in each rock was investigated as a natural analogue for some HLW elements to identify the nuclide migration pathways and significant sites of rock/radionuclide interaction. Secondly, Synroc doped with actinides and fission products was used as a source of radionuclides to evaluate the interactions between waste elements and intrusive rocks in a simulated water-saturated repository environment. This integrated approach has identified the mechanisms controlling radionuclide release, migration and retention.

In the natural analogue studies, the application of fission-track micromapping has determined that primary uranium is distributed in the three intrusives as background U in the major rock-forming minerals, and as resistate U in the primary accessory phases. Two modes of redistribution of this uranium are evident; as secondary U in the secondary minerals formed during alteration, and as fracture U associated with the fracture-infilling minerals. The mechanisms for uranium retention are dominated by adsorption and ion exchange. The study of uranium-series disequilibrium has determined that significant radionuclide mobilisation has occurred in the recent past (<1.2 Ma) as a result of groundwater interaction. Disequilibrium between ^{230}Th , ^{234}U and ^{238}U in the three intrusives indicates that fractures form the

dominant nuclide migration pathways and the most significant sites of rock/radionuclide interaction are the secondary and fracture-infilling minerals.

Leach testing of Synroc with the three intrusives was carried out to determine the mechanisms and processes which occur during Synroc/water/granitic host rock interaction. Significant geochemical and mineralogical changes were observed in all three intrusives during leach testing, including loss of crystal structure and formation of surface reaction products. These changes are reflected in a change in the leach solution conditions and may also affect the distribution of radionuclides during leach testing. The presence of the intrusives significantly inhibited the total release of the actinides (Np, Pu and Cm) and the less soluble fission products (Zr, Ce, Nb and Ru) from Synroc, as a result of the change in solution chemistry and nuclide solubility induced by the presence of the granites. Substantial preferential uptake of all radionuclides by specific secondary and fracture infilling minerals (such as sericite, hematite, Fe- and Ti-oxides/hydroxides) in intrusives was also observed, which was controlled by rapid ion exchange, redox reactions, sorption and surface deposition of colloids and pseudocolloids.

These results imply that fractures will form the main pathways for radionuclide mobilisation in the HLW disposal environment, and that the most significant sites for rock/radionuclide interaction will be the secondary and fracture-infilling minerals. Sorptive processes will dominate radionuclide retention and therefore retard migration of these elements away from the waste package into the surrounding near-field geological environment. The geochemical evolution which occurs during rock/water interaction may affect radionuclide release, migration and retention through changes in solution characteristics and sorptive capacity. These qualitative experimental observations of the chemically complex interactions which occur in the predicted repository environment may be used for quantitative predictive modelling in repository assessment.

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Table of Contents

Abstract.....	i
Acknowledgements	iii

SECTION I: INTRODUCTION AND BACKGROUND

Chapter 1: *The Nuclear Waste Disposal Problem*

1.1 Introduction	1:1
1.2 Background	1:2
1.2.1 The Fundamental Problem - Nuclear Waste	1:2
1.3 Geological Disposal of HLW	1:6
1.3.1 The Multiple Barrier Concept.....	1:8
1.3.2 The Geological Barrier	1:10
1.4 Aims and Objectives of this Study.....	1:14

Chapter 2: *Regional Geology*

2.1 Granites in the High-Level Waste Disposal Environment	2:1
2.2 Sampling Techniques and Analytical Methods.....	2:4
2.3 Coles Bay Granite	2:9
2.3.1 Field Relationships.....	2:9
2.3.2 Petrography.....	2:10
2.3.3 Whole Rock Geochemistry	2:15
2.4 Kambalda Granodiorite	2:18
2.4.1 Field Relationships.....	2:18
2.4.2 Petrography.....	2:20
2.4.3 Whole Rock Geochemistry	2:24
2.5 Roxby Downs Granite	2:28
2.5.1 Field Relationships.....	2:28
2.5.2 Petrography.....	2:30
2.5.3 Whole Rock Geochemistry	2:34

SECTION II: THE BEHAVIOUR OF NATURAL URANIUM AND THORIUM IN GRANITES

Chapter 3: *Fission-Track Analysis of Uranium Distribution in Granites*

3.1 Introduction	3:1
3.2 Analytical Methods	3:1
3.2.1 Fission-track Micromapping as an Analytical Tool	3:1

<i>3.3 Results: Fission-track Micromapping</i>	3:4
3.3.1 Background U.....	3:9
3.3.2 Resistate U.....	3:9
3.3.3 Secondary U.....	3:16
3.3.4 Fracture U.....	3:21
<i>3.4 Discussion</i>	3:24
3.4.1 Primary Uranium Distribution.....	3:24
3.4.2 Secondary Uranium Distribution.....	3:26
<i>3.5 Conclusions</i>	3:32

Chapter 4: *The Use of Uranium-Series Disequilibrium Studies to Determine Recent Rock/Groundwater Interactions in Granites*

<i>4.1 Introduction</i>	4:1
4.1.1 Naturally Occurring Radioactive Decay Series - the Uranium-Decay Series.	4:1
4.1.2 Geochemistry of the U-Decay Series and the Occurrence of Radioactive Disequilibrium.....	4:5
4.1.3 Application of U-Series Disequilibrium Studies as a Natural Analogue of the HLW Disposal Environment.	4:8
<i>4.2 Characterisation of U-Series Disequilibrium in Three Australian Granites</i>	4:9
4.2.1 Experimental Methods.....	4:9
4.2.2 Results - U-Series Disequilibrium Analysis	4:10
<i>4.3 Mechanisms for Isotopic Fractionation</i>	4:19
<i>4.4 Implications for HLW Disposal</i>	4:25
<i>4.5 Conclusions</i>	4:26

SECTION III: AN INTEGRATED STUDY OF THE INTERACTION BETWEEN SYNROC AND GRANITE

Chapter 5: *Alternative Wasteforms for HLW Disposal*

<i>5.1 Introduction to Wasteforms — Previous Work</i>	5:1
<i>5.2 Synroc as a Wasteform Alternative</i>	5:2
5.2.1 Chemical Durability.....	5:3
<i>5.3 Comparative Behaviour of Synroc and Borosilicate Glass</i>	5:8

Chapter 6: *The Crystalline HLW Repository Environment*

<i>6.1 Factors affecting Radionuclide Migration in the Crystalline Repository Environment</i>	6:1
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6.2 <i>The Behaviour of Granites as a Potential Disposal Medium</i>	6:3
6.2.1 Experimental Procedure	6:3
6.2.2 Granite Behaviour under Leach Conditions.....	6:4
6.3 <i>Discussion</i>	6:22
6.4 <i>Conclusions</i>	6:25

Chapter 7: *Simulation of Repository Processes I: The Interaction of Granite and Synroc doped with the Actinides*

7.1 <i>Introduction - Previous Work</i>	7:1
7.2 <i>Experimental Techniques</i>	7:3
7.3 <i>Results</i>	7:6
7.3.1 ^{239}Pu	7:8
7.3.2 ^{237}Np	7:15
7.3.3 ^{244}Cm	7:20
7.4 <i>Discussion</i>	7:25
7.4.1 Mobilisation of ^{239}Pu , ^{237}Np and ^{244}Cm from Synroc.....	7:25
7.4.2 Mechanisms for ^{239}Pu , ^{237}Np and ^{244}Cm Distribution.....	7:27
7.5 <i>Conclusions</i>	7:30

Chapter 8: *Simulation of Repository Processes II: The Interaction of Granite and Synroc doped with Mixed Fission Products*

8.1 <i>Introduction - Previous Work</i>	8:1
8.2 <i>Mixed Fission Products</i>	8:4
8.2.1 Experimental Techniques.....	8:4
8.2.2 Results.....	8:6
8.3 <i>Uptake of ^{134}Cs and ^{90}Sr from Radionuclide-Doped Solution</i>	8:17
8.3.1 Experimental Techniques.....	8:17
8.3.2 Results.....	8:19
8.4 <i>Discussion</i>	8:30
8.4.1 Solution Characteristics	8:30
8.4.2 Mechanisms for Retention of Radionuclides by Granites	8:32
8.5 <i>Conclusions</i>	8:35

SECTION IV: IMPLICATIONS AND CONCLUSIONS

Chapter 9: *Implications for HLW Disposal*

<i>9.1 The Repository Environment - an Integrated Study</i>	9:1
<i>9.2 Natural Analogues — What information can they provide?</i>	9:4
<i>9.3 The Three Component Simulation of the Repository Environment Processes</i>	9:6
<i>9.4 Repository Assessment and Predictive Model Validation</i>	9:10
<i>9.5 Granites as a Candidate Repository Medium</i>	9:13
<i>9.6 Future Research arising from this Study</i>	9:15

Chapter 10: *Conclusions*

<i>10.1 Conclusions arising from this Study</i>	10:1
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SECTION V: REFERENCES AND APPENDICES

References	R:1
Appendices	A:1